Design of an Ultrafast Frequency Doubling Photonic Device *

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Ultrashort pulses complicate the frequency conversion in a nonlinear crystal, where group velocity mismatch becomes the main obstacle due to dispersion. We present a design for group velocity compensated second harmonic generation in a modulated nonlinear structure, embedded in a liquid crystal box. In this structure, nonlinear crystals act as sources of signal and liquid crystals compensate for group velocity mismatch originating from nonlinear crystals. There are the advantages of the flexible, controllable birefringence of liquid crystals. Meanwhile, a method calculating the parameters of this type of structure is presented. To make it clear, an example is provided. Furthermore, the structure can also be shaped as a waveguide to support integration into other optical devices, applicable to all-optical processing systems.

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The rapid development of optical communications requires faster optical processing devices and shorter optical pulses transmitted in fibers. Ultrashort pulses are also preferable in high speed optical information processing and computing, where their accumulated dispersion in fibers is not too severe to be ignored. In various designs, the second-order nonlinearity in devices is commonly used for realizing related all-optical technologies such as switchings, logic gates and wavelength conversion devices.[1–3] When the sum/difference frequency generation and the second harmonic generation (SHG) are involved, there are three kinds of devices which are mostly applied now, i.e., periodically poled ferroelectric crystals,[4] semiconductors (mixed GaAs series) and traditional nonlinear crystals (e.g. β-BBO, CLBO). In the first class, typically, periodically poled lithium niobate is a representative which possesses a large nonlinearity (at least 20 times larger than β-BBO), thus is greatly applied in the 1.3 and 1.5 μm bands with the quasi-phase matching technique.[1–6] In the same transmission band, mixed GaAs materials (typically AlGaAs), always structured as superlattice waveguides, have distinct functionality and advantages in nonlinearity and integration.[7–9] However, they in general carry problems in growth processes, such as incurring large absorbing and light scattering, and are required to meet phase matching (PM) condition, which renders additional technical complexity.[10] The common bulk nonlinear crystals with birefringent PM are the simplest frequency converters with highest optical uniformity and damage threshold, while with smaller nonlinearity. The main problem in the three classes is the group velocity mismatch (GVM) in coupling processes, which broadens pulses, shortens converter's bandwidth and finally loses the advantage of ultrashort pulses in all-optical systems. Moreover, it is difficult to extend periodically poled ferroelectric crystals and semiconductors to the high frequency band (800–900 nm). We present one design composed of nonlinear crystals and liquid crystals in this work to overcome the difficulty by use of birefringent PM, which can be used in a broad communication frequency band. Compared with our previous design,[11] this design has the advantages of controllable dispersion, easy machining, etc.

Frequency conversion in almost all the nonlinear crystals having access to birefringence PM is restricted by their dispersions. The dispersion renders limited spectral acceptance and dispersive group velocity which leads to the GVM for ultrashort pulses. The structuring technique can be used to reach PM and even surmount GVM problems, such as multi-crystal configurations which equivalently turn linear and nonlinear susceptibilities into some patterned forms in the beam propagation direction.[12–17] A periodic modulation which engineers the birefringence can nearly remove the GVM by walk-off compensation,[14,15] and its efficiency will gain step-by-step as shown in Fig. 1. In this structure, the total efficiency will be lower than that of a pure bulk nonlinear crystal if the GVM is ignored, as shown in Fig. 1. Comparatively, we can use a reduced nonlinear coefficient $\sigma_{\mathrm{R}} = \sigma(d_1/\Lambda)^2$ to describe the ability of the periodic structure (σ is the coefficient of a nonlinear crystal, d_1 is the thickness of each nonlinear layer, and d_2 is that of a compensat-

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ing layer). The method performed well in the periodic walk-off compensation technique in the multi-crystal configuration,[15] which nevertheless, is impossible for miniature design in integrated photonic devices. The following design transplants the idea into a special controllable material.

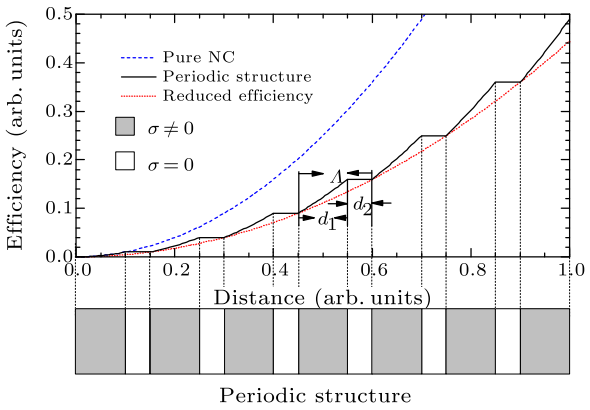

Fig. 1. SHG efficiency of a periodic nonlinear structure compared with that of the pure nonlinear crystal where the period is $\Lambda = d_1 + d_2$, the filled squares denote the nonlinear material, and the hollow ones represent compensating layers which have a larger birefringence but a disregarded nonlinearity.[18,19] The simulation uses the undepleted pump approximation.

According to the compensation condition,[15] it requires

$$\Delta u_1(\theta_1)d_1 + \Delta u_2(\theta_2)d_2 = 0, \tag{1}$$

where $\Delta u_j = 1/u_{\mathrm{S}j} - 1/u_{\mathrm{F}j}$ is the inverse group velocity mismatch with $u_{\mathrm{F}j}$ being the group velocity of the fundamental pulses, $u_{\mathrm{S}j}$ being the second harmonic pulse, $j = 1$ for the nonlinear layers, $j = 2$ for the compensating layers, θ_1 is the PM angle (fixed for a given pumping wavelength), and θ_2 is the angle of the harmonic wave vector made with the optical axis of the compensating layers. This equation makes the fast one between fundamental and harmonic pulses be slowed in the compensating layers, thus the overall GVM vanishes. Moreover, an ideal SHG also needs PM condition: $\Delta k_1 = 2k_{\mathrm{F}1} - k_{\mathrm{S}1} = 0$, where k represents the wave constant, the subscripts have the same meanings as the above. In other words, $2k_{\mathrm{F}1}$ also represents the wave constant of the nonlinear polarization (a square of the fundamental field). To guarantee an effective SHG, the compensating crystal should be phase matched as well. It should be

$$\Delta k_2(\theta_2)d_2 = 2n\pi, \quad n \in \mathbb{Z}, \tag{2}$$

which cancels the phase difference between the harmonic and the nonlinear polarization when they pass through the compensating layers, otherwise it will destroy the total SHG despite PM condition in nonlinear layers being satisfied. Equation (2) promises an ideal coherence among all the harmonic pulses generated in different nonlinear layers. By adjusting the segment ratio of d_2/d_1, Eq. (2) can be satisfied with various values of n. Bear in mind that Eqs. (1) and (2) deeply depend on the propagation direction in the compensating layers.

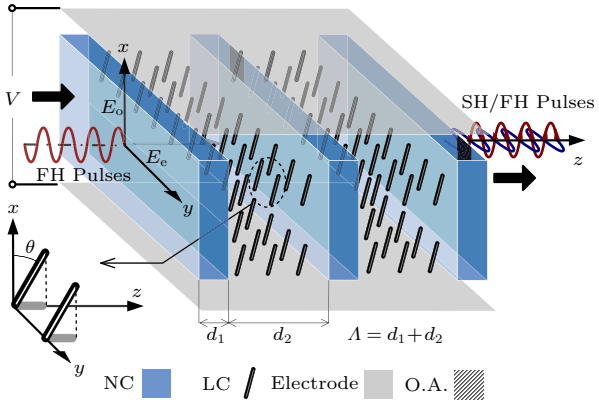

Fig. 2. Periodic LC embedded nonlinear structure as a walk-off compensating type-I SHG device, where the nonlinear crystal is at its PM stage.

We can take one nonlinear material as the SHG source, and choose a suitable liquid crystal (LC) to compensate for GVM by its adjustable birefringence. A simplified design is shown in Fig. 2, where the nonlinear crystal (labeled as NC) is not only the source of SHG but also the core of the structure. One kind of LC with appreciable birefringence can be put in the open grooves in the crystal bolster, and then sealed as an LC box. Electrodes can be produced on the top and bottom of the box with a lithographic technique. Certainly, we need some LC alignment techniques to ensure the orientation of LC molecules, which are practically mature in LC display fabrication processes. The direction of electrode depends on the optical properties of the LC. In Fig. 2, we endow the nonlinear crystal with a negative uniaxiality (the orientation should be redirected for a positive crystal) and the LC as a positive structure in terms of a general optical property of nematic LCs.[20] Thus the ordinary (o) and extraordinary (e) lights will be vertically and horizontally polarized respectively in the nonlinear crystal for the type-I (oo-e) configuration, but are reversed as ee-o when into the LC in accordance with the orientation of the director. In this case, conditions Eqs. (1) and (2) should be satisfied. The known electric-control birefringence effect provides a feasible method to adjust the refractive index (e light) of the nematic LC. Within 5 V over its threshold, the birefringence of LC can be maximally altered in a commonly nonlinear variation mode, which should be identified by experiment for a specified LC material. This electric-control mechanism comes from the Frederiks transition induced by distortion free energy. Further discussions concerning LC dynamics about temperature depen-

dence and boundary effects are not intended here for concision. This LC embedded structure features in the tuning ability of birefringence readily by applying voltage. Accordingly, it is convenient in practice to find an optimal working point.

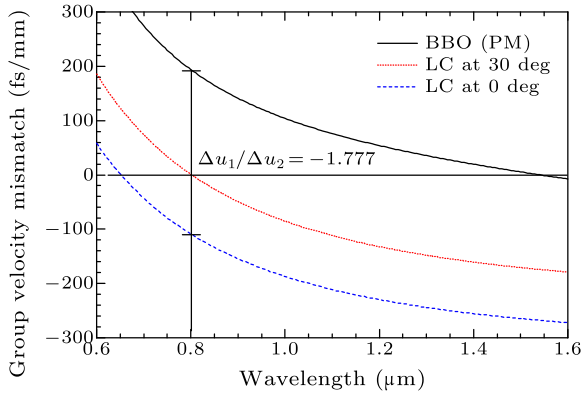

Fig. 3. Inverse group velocity mismatches for BBO and MLC-9200-100, where $\theta_1 = \theta_1(\lambda)$ varying with wavelength is the PM angle. The two curves of MLC-9200-100 are taken at two special angles $\theta_2 = 0$ and 30°, respectively.

For a detailed illustration, we consider an SHG process pumped by a mode-locked Ti:sapphire laser with a central wavelength of 800 nm, pulse width of $\tau = 100$ fs (full width at half maximum), and repetition rate of 80 MHz at room temperature 25℃. Here we use the typical crystal beta-borate (β-BBO) as the nonlinear structure. Its dispersion leads to 194 fs/mm of Δu_1, which presents a walk-off length ($L_{\rm W} = \tau/|\Delta u_1|$) of 0.51 mm. Thus the thickness of a single BBO layer should be less than this value. The typical LC material, MLC-9200-100, is chosen as the LC box, which is commonly used in display panels.[21] MLC-9200-100 possesses the birefringence $\Delta n \approx 0.11$ (positive uniaxiality) and carries the absorption edge at $\lambda_{\rm cut} \approx 300$ nm, which allows for an effective frequency conversion.[21] Certainly, we can find many LCs with an even larger birefringence, which gives us a great freedom to choose in practice. In Fig. 3, we present the inverse group velocity mismatch of BBO and LC, where the refractive indices come from Refs. [21,22], respectively. The maximum of $-\Delta u_2$ is at $\theta_2 = 0$ and it yields a minimum of $d_2/d_1 = 1.777$ according to Eq. (1). However, the ratio increases with θ_2 and approaches to infinity at about $\theta_2 = 30°$, where the structure is unable to compensate for the GVM in BBO.

By a direct computation, we obtain some admitted segment lengths and reduced nonlinear coefficients, as shown in Fig. 4. The compensation condition (1) infers that the segment length d_2 is related to the BBO gap d_1 by a scale factor. The same parameter d_2 in Eq. (2), nevertheless, does not depend on the gap and should be taken at some special values for coherent output. The BBO layer thicknesses in the two cases all rely on the compensating angle of the LC. For the argument d_1, the two variation modes of d_2 from Eqs. (1) and (2) are exhibited in Fig. 4(a) (sampled for $n = 26$ at three angles) and the crossing points thereof just give the solutions of the simultaneous equations. Therefore, an implicit function d_2 of variable d_1 can be determined for a given number n, which is a connection of all the crossing points like A, B, C in the figure. Likewise, another function relation of θ_2 to d_1 can be solved. Thus the two equations also produce an optimal orientation of the LC via a substitution of the expression d_2 from Eq. (1) into Eq. (2)

$$\theta_2 = \theta_2(d_1, n), \quad n \in \mathbb{Z}, \tag{3}$$

which is demonstrated in Fig. 4(b).

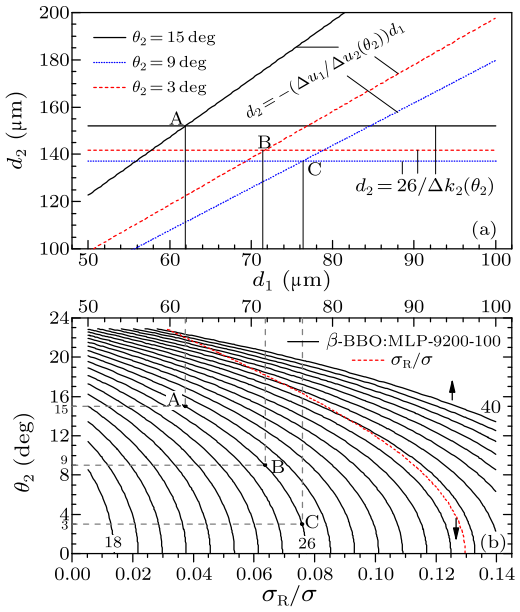

Fig. 4. Demonstration of permitted segment lengths (a) derived from Eqs. (1) and (2) and GVM compensating angles (b) of the periodic LC embedded structure with the ratio curves of the reduced nonlinear coefficients. There are sampled points A, B, C at $n = 26$.

On the other hand, LCs must be suitable for the device fabrication in transparence range, temperature sensibility (needs control in LCs), scattering due to refractive index difference (weak), chemical stability with the source crystal and so on. With a fixed parameter n, a combination of Eqs. (1) and (2) will determine d_2 and Λ if d_1 is given. A smaller angle produces a higher efficiency due to the positivity of the LC. Furthermore, a large parameter n is required to catch a higher nonlinearity. Even though Eqs. (1) and (2) are efficient to produce an effective SHG, they are not the only conditions which must be satisfied. Equation (2) can be extended to a more general one,

$$\Delta k_1 d_1 + \Delta k_2(\theta_2) d_2 = 2n\pi, \tag{4}$$

which relaxes the PM condition (2), but has a lower reduced nonlinear coefficient. Then, any nonlinear crys-

tal unable to meet PM condition through birefringence is possible to be utilized in the modulated structure.

Naturally, the above design can be applied to other nonlinear materials under the same mechanism. For example, the design can be applied to the popular periodic polarized $LiNbO_3$ (ee-e PM type) to use its high nonlinearity. In this case, the LC unit should be divided into two parts, the first of that, aligned to another direction, acts as $\lambda/2$ plate to transfer the ee polarization to the oe type (as that in Ref. [22]), then the second part compensates for the mismatch.

In summary, one kind of periodical modulated nonlinear structure has been proposed to realize SHG of ultrashort pulses without pulse broadening, which, as a wavelength conversion device, can be applied in a large communication band with suitable nonlinear materials. The basic principle is to use birefringent materials to compensate for the GVM induced in an SHG process. The micromachined nonlinear elements can be integrated into an optical system as waveguide structures.

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